Contribution of the surface dipole to deformation of superconductors

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The interaction of the ionic lattice with the superconducting condensate is treated in terms of the electrostatic force in superconductors. It is shown that the surface dipole supplies the force responsible for the volume difference of the normal and superconducting states. Assuming this mechanism, we argue that the usual parametrization of the theory of deformable superconductors should be revisited.

I. INTRODUCTION

The theory of deformable superconductors deals either with effects of the atomic lattice deformations on the superconducting condensate or with deformations of the crystal lattice caused by the inhomogeneous superconducting condensate. For example, a lattice deformation around a dislocation pins a vortex.[1,2] On the other hand, the forces generated by supercurrents contribute to the magnetostriction[3–5] and the condensate depletion at the vortex core deforms the atomic lattice so strongly that a significant renormalization of the vortex mass has been predicted.[6–9] In some cases, one cannot say which of the two effects dominates. This happens, for instance, if the ionic lattice deformation influences the structure or orientation of the Abrikosov vortex lattice.[10]

The free energy describing deformable superconductors has to include at least three parts. The first part is the elastic energy of deformations. Its structure and parametrization have well been established for a long time.[11] The second part is the magnetic energy and the energy of superconducting condensation. This part can be covered on different levels. Here, we will refer to the Ginzburg-Landau (GL) theory[12] that was employed in the majority of the above mentioned studies. The third part is a cross term which describes the mutual effect of deformations and the condensate. In this paper, we focus on this interaction term.

In the phenomenological approach put forward by Kramer and Bauer,[1] the interaction of the lattice and the condensate is described by the local product of the atomic lattice density with the density of superconducting electrons. The strength of this interaction is conveniently fitted to the volume difference of the normal and superconducting states.

This local interaction is justified only if the system remains charge neutral.[11] Superconductors, however, have a small charge transfer which results in the so-called Bernoulli potential.[13–15] Moreover, the superconducting gap modifies the surface dipole.[16] The former effect has been already discussed in Ref. 17, where nonlocal corrections to the theory of deformable superconductors were derived. To our knowledge, the latter effect has not been considered so far. This effect of the surface dipole is the major focus of interest in the present paper.

Before we specify the paper content, let us take a look at how the problem in question meets the general assumptions of the theory of deformable superconductors. If the lattice is modeled by an isotropic deformable medium and the interaction is assumed to be local, the interaction energy of Kramer and Bauer is the only one compatible with the system symmetry. Indeed, the condensate density is a scalar which can interact only with another scalar. The only isotropic scalar quantity linear in the deformation is the trace of the strain tensor, which is proportional to the ionic density. Of course, one can construct more elaborate scalars within nonlinear terms but these are higher-order corrections.

Two modifications are at hand. First, one can take into account that the real atomic lattice is never isotropic. Even in simplest lattices of elementary metals, the shear rigidity depends on the orientation of the deformation with respect to the crystal axes. In the anisotropic crystal, there are two scalar quantities linear in the shear deformation. A corresponding anisotropic generalization of the interaction between the condensate and the ionic lattice has been discussed by Kogan *et al.*[10]

Second, one can go beyond the local approximation. The reason for such a step was already mentioned—the interaction in the local approximation is justified only for neutral systems.[11] The superconducting condensate, however, drives the system out of neutrality inducing the electrostatic potential known as the Bernoulli potential.[18,19] The nonlocal interaction mediated by the Bernoulli potential has the form of the quantum kinetic energy and within the GL theory, it can be recast into a nonlinear but local interaction.[17]

Another contribution to the charge transfer induced by the condensate is the surface dipole.[16] While all the above mentioned interactions result in a force density acting in the bulk of the crystal, the surface dipole yields a force that acts as an external pressure imposed on the surface. As far as we know, surface contributions to the theory of deformable superconductors have never been discussed. In this paper, we shall fill this gap.

The paper is organized as follows. In Sec. II, we show that the surface dipole determines changes of the crystal volume during its transition from the normal to the superconducting state. To this end, we first introduce the basic concept in Sec. II A and derive the coefficient of the local

interaction from the pressure dependence of the condensation energy at zero temperature in Sec. II B. The result is compared with the force due to the surface dipole in Sec. II C. In Sec. III, we introduce the interaction mediated by the Bernoulli potential. We first derive a formula for the coefficient of the local interaction. In Secs. III A and III B, we evaluate the interaction coefficient for moderately strong and weak coupling superconductors. In Sec. IV, we discuss differences and conclude.

II. LOCAL APPROACH

In their pioneering study, Kramer and Bauer[1] proposed to deduce the interaction strength from the pressure dependence of the critical magnetic field B_c. Since experimental data for this parametrization are conveniently found in literature, this approach has been employed by other authors too.

In this section, we provide a derivation of the local interaction of Kramer and Bauer within the GL picture of the superconductor. The presented approach is based on papers by Šimánek[6] and Hake.[20]

A. Phenomenological force

The volume of a metal changes at the phase transition from V_n in the normal state to V_s in the superconducting state.[20,21] This change is described by a relative change α of the volume defined as

$$V_n - V_s = \alpha V_s. \qquad (1)$$

Since the number of atoms N conserves, we can write the same relation for the volume per atom $v = V/N$, i.e., $v_n - v_s = \alpha v_s$.

If the volume per atom becomes inhomogeneous, the crystal has regions requiring different distances of neighboring atoms. This leads to internal stresses which can be expressed via an effective force density[11]

$$\mathbf{F}_{\mathrm{ph}} = K \nabla \alpha, \qquad (2)$$

where K is the modulus of hydrostatic compression or simply the bulk modulus. It is defined as the inverse of the relative volume change with respect to the pressure

$$\frac{1}{K} = -\frac{1}{V} \frac{\partial V}{\partial p}. \qquad (3)$$

The temperature dependence of α is similar to the temperature dependence of the superconducting fraction

$$\frac{\alpha}{\alpha_0} \approx \frac{|\psi|^2}{|\psi_0|^2}, \qquad (4)$$

where the subscript zero denotes the zero temperature value. Šimánek[6,7] and Coffey[8,9] use the BCS gap Δ,

$$\frac{\alpha}{\alpha_0} \approx \frac{|\Delta|^2}{|\Delta_0|^2}, \qquad (5)$$

while other authors prefer the GL function ψ. We will restrict our attention to the vicinity of the critical temperature, where

both forms are equivalent since the BCS gap and the GL function are linearly proportional to each other.[22]

Relation (4) is the central approximation in the phenomenological theory of deformable superconductors. Assuming that in the normal state the system is homogeneous, α_0 and ψ_0 are constants. With the GL function normalized to the density of pairable electrons $2|\psi_0|^2 = n$, we obtain

$$\mathbf{F}_{\mathrm{ph}} = \frac{2}{n} \alpha_0 K \nabla |\psi|^2, \qquad (6)$$

which we will use in our discussion.

B. Difference of normal and superconducting volume

Now, we link the relative change of the volume α to the pressure dependence of the condensation energy $\varepsilon_{\mathrm{con}}$. We follow the derivation of Hake.[20]

The volume of the sample is the pressure derivative at fixed temperature of the Gibbs free energy

$$V = \left(\frac{\partial G}{\partial p} \right)_T. \qquad (7)$$

At zero magnetic field and zero temperature, the free energy of the normal state G_n is higher than the superconducting free energy G_s by the condensation energy

$$G_n - G_s = V_s \varepsilon_{\mathrm{con}}. \qquad (8)$$

Due to the complete expulsion of the magnetic field from type-I superconductors, the condensation energy is conveniently observed via the critical magnetic field at zero temperature, B_0,

$$\varepsilon_{\mathrm{con}} = \frac{B_0^2}{2\mu_0}. \qquad (9)$$

In his study, Hake expresses all thermodynamical relations exclusively in terms of the critical magnetic field. Here, we prefer to use the condensation energy.

From the pressure derivative of relation (8), it follows that

$$V_n - V_s = V_s \frac{\partial \varepsilon_{\mathrm{con}}}{\partial p} + \varepsilon_{\mathrm{con}} \frac{\partial V_s}{\partial p}. \qquad (10)$$

Comparing the thermodynamical relation [Eq. (10)] with the definition [Eq. (1)], we obtain the coefficient α_0 in terms of the condensation energy

$$\alpha_0 = \frac{\partial \varepsilon_{\mathrm{con}}}{\partial p} + \varepsilon_{\mathrm{con}} \frac{1}{V_s} \frac{\partial V_s}{\partial p}. \qquad (11)$$

In terms of the bulk modulus [Eq. (3)], we have

$$\alpha_0 = \frac{\partial \varepsilon_{\mathrm{con}}}{\partial p} - \frac{\varepsilon_{\mathrm{con}}}{K}. \qquad (12)$$

The force [Eq. (6)] depends on the product $\alpha_0 K$; it is thus advantageous to introduce the inverse bulk modulus also into the first term of Eq. (12). For simplicity, we consider hydrostatic pressure and conventional superconductors with isotropic structure. In this case, we can express the pressure de-

pendence of the density of the condensation energy ε_{con} via its dependence on the electron density,

$$\frac{\partial \varepsilon_{\text{con}}}{\partial p} = \frac{\partial \varepsilon_{\text{con}}}{\partial n} \frac{\partial n}{\partial p}. \tag{13}$$

Since the number of electrons N does not change, we can express the bulk compressibility via the change of the density $n = N/V$ as

$$\frac{1}{K} = \frac{1}{n} \frac{\partial n}{\partial p}. \tag{14}$$

Using relation (13) and the bulk modulus [Eq. (14)] in Eq. (12), we get

$$\alpha_0 K = n \frac{\partial \varepsilon_{\text{con}}}{\partial n} - \varepsilon_{\text{con}}. \tag{15}$$

The density derivative is taken under the condition of charge neutrality, i.e., the atomic lattice density changes with the electron density.

The phenomenological force [Eq. (6)] according to relation (15) thus reads

$$\mathbf{F}_{\text{ph}} = 2\left(\frac{\partial \varepsilon_{\text{con}}}{\partial n} - \frac{\varepsilon_{\text{con}}}{n} \right) \nabla |\psi|^2. \tag{16}$$

As far as we know, in metals, the derivative is the dominant part, $\frac{\partial \varepsilon_{\text{con}}}{\partial n} \gg \frac{\varepsilon_{\text{con}}}{n}$. Many authors keep only this dominant term neglecting the other for simplicity.

C. Compression via the surface dipole

Now, we show that the volume change can be described as a compression of the sample by a force created by the surface dipole. This relation makes use of the parameter α_0 for interactions deep in the bulk questionable.

At the surface of a metal, the electrostatic potential rises by few volts from its vacuum value to the value deep in the metal.[23] This increase is spread partly outside the region occupied by ions, typically on the scale of the tunneling length of electrons in the potential barrier given by the work function. A part of the barrier is located inside the metal on the scale of the Thomas-Fermi screening length. Both scales are of the order of angstroms, making the potential step very sharp. This sharp step is called the surface dipole.

The surface dipole naturally depends on the temperature. Moreover, when the metal undergoes a transition to the superconducting state, the temperature dependence of the surface dipole changes. Briefly, the superconducting condensate affects the surface dipole.[16] Let us denote this additional potential near the surface as φ_T.

The amplitude $\varphi_T(0) - \varphi_T(\infty)$ of the additional potential step follows from the Budd-Vannimenus theorem as[16]

$$\rho_{\text{lat}}[\varphi_T(0) - \varphi_T(\infty)] = f - n\frac{\partial f}{\partial n}, \tag{17}$$

where $f = f_s - f_n$ is the free-energy density by which the superconducting state differs from the normal state and ρ_{lat} is the charge density of the ionic lattice. We assume a superconductor which fills the half-space $x > 0$.

The additional potential exerts an electrostatic force density on the ionic lattice,

$$\mathbf{F}_T = -\rho_{\text{lat}} \nabla \varphi_T. \tag{18}$$

The integral of this force density across the surface region corresponds to an effective pressure on the lattice,

$$p_T = \int_0^\infty dx F_T^x = \rho_{\text{lat}}[\varphi_T(0) - \varphi_T(\infty)], \tag{19}$$

which changes the volume of the crystal by

$$\tilde{\alpha}V = \frac{\partial V}{\partial p}p_T. \tag{20}$$

Clearly, the surface dipole contributes to the relative change of the volume $\tilde{\alpha}$. From Eq. (20), we find

$$\tilde{\alpha} = \frac{1}{K}\rho_{\text{lat}}[\varphi_T(0) - \varphi_T(\infty)]. \tag{21}$$

At zero temperature $f = -\varepsilon_{\text{con}}$, therefore from Eq. (17), it follows that

$$\rho_{\text{lat}}[\varphi_0(0) - \varphi_0(\infty)] = n\frac{\partial \varepsilon_{\text{con}}}{\partial n} - \varepsilon_{\text{con}}. \tag{22}$$

The dipole-induced volume change at zero temperature thus reads

$$\tilde{\alpha}_0 = \frac{1}{K}\left(n\frac{\partial \varepsilon_{\text{con}}}{\partial n} - \varepsilon_{\text{con}} \right). \tag{23}$$

Comparing Eq. (23) with Eq. (15), one can see that the volume change is fully induced by the surface dipole

$$\alpha_0 = \tilde{\alpha}_0. \tag{24}$$

The fact that the volume change is caused by the surface dipole shows that one should be cautious using the relative change of the volume α_0 as a coefficient of the interaction between the ionic lattice and the condensate.

Studies of the electrostatic potential in superconductors have shown that the bulk and surface potentials are of different nature and are covered by distinct theories. We note that these theories are experimentally verified. The surface potential including the surface dipole has been observed by Morris and Brown via the Kelvin capacitive pickup.[16,19] The internal charge transfer caused by the bulk electrostatic potential in the vortex core has been observed via the nuclear magnetic resonance by Kumagai et al.,[24] see also Lipavský et al.[25]

III. ELECTROSTATIC FORCE ON IONS

According to the Hellmann-Feynman theorem, electrons act on ions exclusively via the electrostatic force.[31] In this spirit, we expect the force density to be of electrostatic nature,

$$\mathbf{F}_{\text{el}} = -\rho_{\text{lat}} \nabla \varphi, \tag{25}$$

where φ is the electrostatic potential created by the superconducting electrons which is conveniently derived follow-

ing the approach of Rickayzen.[15] Since the system is in equilibrium, the Gibbs electrochemical potential μ for electrons is constant all over the sample. It is locally defined from the density of free energy f as

$$\mu = e\varphi + \frac{\partial f}{\partial n}. \tag{26}$$

Following the customary choice in the theory of superconductivity, we set the electrochemical potential to zero, $\mu=0$; therefore,

$$\varphi = -\frac{1}{e}\frac{\partial f}{\partial n}. \tag{27}$$

The theory of the electrostatic potential has been derived under the assumption that the ion lattice is stiff and its deformation is not included. A combination of both effects has not been studied so far; therefore, it is not yet clear how the density derivative in Eq. (27) is modified by atomic lattice deformations. For simplicity, we assume that the density derivatives in Eqs. (27) and (17) are the same. This is the case if the pairing interaction has a purely electronic nature so that the atomic lattice density has no effect on the condensation energy.

Formula (27) is quite general. It has been employed by Rickayzen to evaluate the Bernoulli potential in superconductors using the London theory supplemented by the phenomenological temperature dependence of the superconducting density. In the same paper,[15] Rickayzen has used formula (27) with the BCS free energy and recovered the result of Adkins and Waldram.[14]

We use the free-energy density in the GL approximation,

$$f = a(T-T_c)|\psi|^2 + \frac{1}{2}b|\psi|^4 + \frac{1}{2m^*}|(-i\hbar\,\nabla - e^*\mathbf{A})\psi|^2$$

$$+ \frac{1}{2\mu_0}|\mathbf{B}_a - \nabla \times \mathbf{A}|^2, \tag{28}$$

where $\mathbf{A}$ is the vector potential and $\mathbf{B}_a$ is the applied magnetic field. The reader not familiar with the GL theory is referred to the textbook[26] of Tinkham.

The magnetic free energy [the last term of Eq. (28)] does not depend on the electron density. For simplicity, we also assume that the Cooper pair mass m^* is independent of this density. Since the GL wave function ψ and the vector potential $\mathbf{A}$ are independent variational fields, the density derivative of the free energy yields the electrostatic potential

$$\varphi = \frac{a}{e}\frac{\partial T_c}{\partial n}|\psi|^2 - \frac{T-T_c}{e}\frac{\partial a}{\partial n}|\psi|^2 - \frac{1}{2e}\frac{\partial b}{\partial n}|\psi|^4. \tag{29}$$

The nonlocal corrections discussed in Ref. 17 are hidden in the second and third terms of Eq. (29). They can be made explicit using relations for material parameters a and b, e.g., Eq. (33), and the GL equation, which couples nonlocal and nonlinear contributions.

Here, we restrict our attention to the close vicinity of the critical temperature, where all nonlocal and nonlinear contributions can be neglected. Indeed, for $T \to T_c$, the GL wave function vanishes as $|\psi|^2 \propto T_c - T$. In lowest order in $T-T_c$,

we can neglect the second and the quartic terms so that relation (29) simplifies to

$$\varphi = \frac{a}{e}\frac{\partial T_c}{\partial n}|\psi|^2. \tag{30}$$

Now, we are ready to evaluate the force acting on the ionic lattice. The electrostatic force density [Eq. (25)] with the electrostatic potential [Eq. (30)] reads

$$\mathbf{F}_{el} = -\frac{\rho_{lat}}{e}\nabla\left(a\frac{\partial T_c}{\partial n}|\psi|^2\right). \tag{31}$$

To first order in $T_c - T$, gradients of material parameters a and $\partial T_c/\partial n$ do not contribute, i.e.,

$$\mathbf{F}_{el} = an\frac{\partial T_c}{\partial n}\nabla|\psi|^2. \tag{32}$$

We have used $\rho_{lat}=-en$ demanded by the local charge neutrality.

With nonlocal and nonlinear corrections neglected, the electrostatic force [Eq. (32)] like the phenomenological force [Eq. (16)] is proportional to the gradient of the superconducting density $|\psi|^2$. Our next aim is to compare the electrostatic coefficient $an\frac{\partial T_c}{\partial n}$ with its phenomenological precursor $2\left(\frac{\partial\varepsilon_{con}}{\partial n} - \frac{\varepsilon_{con}}{n}\right)$. We will show that the relative values of these coefficients depend on the strength of the pairing interaction.

A. Superconductors with moderately strong coupling

To be able to compare the electrostatic force density [Eq. (32)] with the phenomenological force density [Eq. (6)], we need the GL coefficient a as a function of the electron density n. For metals such as niobium or lead, it is possible to use the asymptotic form of the two-fluid free energy of Gorter and Casimir[27] giving[28]

$$a = a_{GC} = \frac{\gamma T_c}{n}. \tag{33}$$

Here, γ is the linear coefficient of the specific heat.

The critical temperature T_c and the critical magnetic field B_0 at zero temperature are linked via the condensation energy. The two-fluid model yields[29]

$$\frac{1}{4}\gamma T_c^2 = \frac{B_0^2}{2\mu_0}. \tag{34}$$

Within this approximation, the electrostatic force density [Eq. (32)] reads

$$\mathbf{F}_{el}^{GC} = 2\left(\frac{\partial\varepsilon_{con}}{\partial n} - \frac{1}{4}T_c^2\frac{\partial\gamma}{\partial n}\right)\nabla|\psi|^2. \tag{35}$$

One can see that this is similar but not identical to the phenomenological force density [Eq. (16)]. In particular, the same dominant term $\propto\partial\varepsilon_{con}/\partial n$ results from both approaches.

We note that Šimánek and other authors use the approximation $\left|\frac{\partial\varepsilon_{con}}{\partial n}\right| \gg \left|\frac{\varepsilon_{con}}{n}\right|$, i.e., they consider only the derivative in their formulas. Within this accuracy, both formulas are equivalent.

B. Superconductors with weak coupling

Metals such as aluminum have weak electron-phonon coupling and one can use the BCS relations. This approximation results in a slightly different electrostatic force.

From the BCS theory, Gor'kov has obtained the parameters of the GL theory.[22] The linear GL coefficient reads

$$a = a_{\mathrm{BCS}} = \frac{6\pi^2 k_B T_c}{7\zeta(3)E_F}, \qquad (36)$$

where E_F is the Fermi energy. The Riemann zeta function has the value $\zeta(3) = 1.202$.

The BCS and the Gorter and Casimir approximations of a can be related within the free electron model. The electron density determines the Fermi vector $k_F = (3\pi^2 n)^{1/3}$ in terms of which $E_F = \hbar^2 k_F^2/2m$. We also use $\gamma = (2/3)\pi^2 k_B^2 N_0$, where $N_0 = (1/4\pi^2)(2m/\hbar^2)k_F$ is the single-spin density of states. Combining these relations, one finds that both values differ by a numerical factor

$$a_{\mathrm{BCS}} = \frac{12}{7\zeta(3)} a_{\mathrm{GC}} = 1.43 a_{\mathrm{GC}}. \qquad (37)$$

The BCS relation connecting the critical temperature with the condensation energy defined via the magnetic field yields another numerical factor

$$0.947 \frac{1}{4}\gamma T_c^2 = \frac{B_0^2}{2\mu_0}. \qquad (38)$$

In the two-fluid model, we find $T_c^{\mathrm{GC}} = B_0\sqrt{2\gamma/\mu_0}$. The correction

$$T_c^{\mathrm{BCS}} = \frac{T_c^{\mathrm{GC}}}{\sqrt{0.947}} = 1.028 T_c^{\mathrm{GC}} \qquad (39)$$

is by an order of magnitude less important than the factor 1.43 from relation (37), however.

From Eq. (38), we obtain T_c in terms of B_0, which we use in the force density [Eq. (32)]. With the BCS relation [Eq. (36)], the electrostatic force density [Eq. (32)] reads

$$\mathbf{F}_{\mathrm{el}}^{\mathrm{BCS}} = \frac{24}{7\zeta(3)}\left(\frac{1}{0.947}\frac{\partial \varepsilon_{\mathrm{con}}}{\partial n} - \frac{1}{4}T_c^2 \frac{\partial \gamma}{\partial n}\right)\nabla |\psi|^2$$

$$= 3.012\left(\frac{\partial \varepsilon_{\mathrm{con}}}{\partial n} - 0.947\frac{1}{4}T_c^2 \frac{\partial \gamma}{\partial n}\right)\nabla |\psi|^2. \qquad (40)$$

One can see that the dominant contribution is increased by slightly more than 50%, as compared to the phenomenological force density [Eq. (16)] and the Gorter-Casimir approximation [Eq. (35)]. Compared to the Gorter-Casimir approximation, the critical temperature T_c^{GC} is replaced additionally by the BCS value T_c^{BCS}. It will thus be interesting to test the validity of the phenomenological force on materials of rather different coupling strengths.

IV. DISCUSSION

We have shown that the change of the total specimen volume during the superconducting transition can be expressed as a compression caused by the surface dipole. It is thus not justified to parametrize the interaction strength of the condensate deep in the bulk by the coefficient of the volume change.

To derive the interaction strength, we have used the internal electrostatic potential for the atomic lattice deformation in the bulk. The force resulting from the electrostatic potential in the superconductor is similar but not identical to the phenomenological force suggested from the volume change.

For moderately coupled materials well described by the Gorter-Casimir two-fluid model, the phenomenological and the electrostatic forces have identical dominant terms $\propto \partial \varepsilon_{\mathrm{con}}/\partial n$. They differ in the correction terms only. For weakly coupled superconductors covered by the BCS theory, the dominant term is increased by nearly 50%. We have not discussed the strongly coupled superconductors which do not obey any of these limits. One can expect that the dominant term is reduced.

It is a question whether the above derived small differences in the internal forces can be accessed by some of recent experiments. Among the physical phenomena mentioned in the Introduction, the magnetostriction offers the most sensitive experimental technique. Indeed, one can resolve even deformations driven by such small changes in magnetization as those caused by the de Haas–van Alphen effect with relative changes of the susceptibility of the order of 1 in 10^7.[30] A superconductor in the Meissner state is an ideal diamagnet with large but fixed magnetization. Small deviations from ideality appear due to the penetration of the magnetic field into the surface. At the same time, the magnetostriction combines the internal forces with the surface dipole. To separate these two contributions, it will be necessary to analyze how the deformation depends on the sample geometry.

The two approaches compared in this paper represent two extreme models. In the electrostatic approach, all forces are attributed to a mean electric field. In the phenomenological model, the system is treated as locally neutral which implies that all forces are attributed to bonds between ions. We expect that a realistic description requires to combine both approaches.

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[31] One should be cautious about using this general argument. The electronic density is modulated by the ionic potential which results in large local electric fields. Their total force on ions is nontrivial, in particular, when the lattice is strained. Formula (25) omits all local contributions. We do not include these contributions in this paper and leave them for the future work.